

Four-wave mixing, quantum control and compensating losses in doped negative-index photonic metamaterials

Alexander K. Popov,^{1,*} Sergey A. Myslivets,² Thomas F. George,³ and Vladimir M. Shalaev⁴

¹Department of Physics & Astronomy, University of Wisconsin-Stevens Point, Stevens Point, WI 54481, USA

²Institute of Physics of the Russian Academy of Sciences, 660036 Krasnoyarsk, Russian Federation

³Center for Nanoscience, Department of Chemistry & Biochemistry and Department of Physics & Astronomy, University of Missouri-St. Louis, St. Louis, MO 63121, USA

⁴Birck Nanotechnology Center and School of Electrical and Computer Engineering, Purdue University, West Lafayette, IN 47907, USA

*Corresponding author: apopov@uwsp.edu

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The possibility of compensating absorption in negative-index metamaterials (NIMs) doped by resonant nonlinear-optical centers is shown. The role of quantum interference and extraordinary properties of four-wave parametric amplification of counter-propagating electromagnetic waves in NIMs are discussed. © 2008 Optical Society of America

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Negative refractive index metamaterials (NIMs) present a novel class of materials that promise a revolutionary breakthrough in electromagnetics (for review, see, e.g., [1]). Nonlinear optics in such materials remains so far a less developed branch of optics. The possibility of nonlinear electromagnetic responses in such materials attributed to the asymmetry of the voltage-current characteristics of their building blocks was predicted in [2,3]. Recent experimental demonstrations of the exciting opportunities to craft nonlinear optical materials with characteristics exceeding those in natural crystals are reported in [4]. Unique nonlinear-optical (NLO) propagation effects associated with three-wave ($\chi^{(2)}$) coupling in NIMs, as compared with their well known counterparts in natural materials, were revealed in [5–8]. The striking changes in the optical bistability in a layered structure including a NIM layer were shown in [9]. A review of the corresponding theoretical approaches is given in [10]. The most detrimental obstacle toward applications of NIMs is strong absorption that is inherent to this class of materials. The possibility to overcome such obstacles based on three-wave optical parametric amplification (OPA) in NIMs was shown in [7,8]. A great deal of technical problems must be solved, however, in order to match the frequency domains of negative index (NI), strong NLO response and the phase-matching to realize such feasibility. Herewith, we propose and explore an alternative approach associated with *resonant four-wave mixing (FWM) nonlinearities* $\chi^{(3)}$ embedded in NIMs and tailored through quantum control. The possibility of compensating losses and manipulating transparency, refractive index and nonlinear response of the NIM sample with control laser(s) is shown.

The basic idea of the proposed approach is as follows. A slab of NIM is doped by four-level nonlinear centers [Fig. 1(a)] so that the frequency ω_4 falls in the NI do-

main, whereas all the other frequencies are in the positive index domain. Below, we show the feasibility to produce the transparency and amplification for the signal wave at ω_4 controlled by two lasers at ω_1 and ω_3 . These three fields also generate an idler at $\omega_2 = \omega_3 + \omega_1 - \omega_4$, which experiences either ordinary, population-inversion, or Raman amplification provided by the driving field at ω_1 and controlled by another driving field at ω_3 . The amplified idler contributes back to $\omega_4 = \omega_3 + \omega_1 - \omega_2$ through FWM which leads to strongly enhanced OPA. We assume that the wave vectors of all waves, \mathbf{k}_j , are

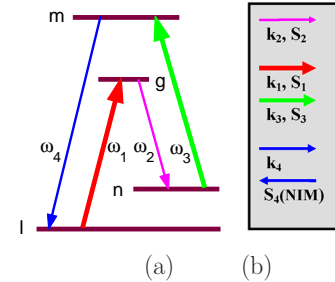


Fig. 1. Scheme of quantum-controlled FWM interaction (a) and coupling geometry (b). ω_4 is signal frequency, ω_2 is idler, and ω_1 and ω_3 are control fields. $n(\omega_4) < 0$.

co-directed, which is required for phase-matching. Since only ω_4 experiences negative refraction and all other frequencies are in the positive index domain, energy flow at ω_4 is counter-directed against other waves. Hence, this signal wave must enter the slab from its opposite side at $z = L$, which is the exit facet for all other waves entering the slab at $z=0$ [Fig.1(b)]. Correspondingly, the signal exits the slab at $z=0$. Such backwardness of the signal wave leads to counterintuitive, distributed feedback-like behavior of the OPA process in NIMs described in [7,8].

The equations describing the coupled waves at ω_4 and ω_2 are as follows

$$da_4/dz = -i\gamma_4^{(3)}a_2^*\exp[i\Delta kz] + (\alpha_4/2)a_4, \quad (1)$$

$$da_2/dz = i\gamma_2^{(3)}a_4^*\exp[i\Delta kz] - (\alpha_2/2)a_2. \quad (2)$$

Here $\gamma_{2,4}^{(3)} = (\sqrt{\omega_4\omega_2}/\sqrt{\epsilon_4\epsilon_2/\mu_4\mu_2})(4\pi/c)\chi_{2,4}^{(3)}E_1E_3$ are NLO coupling coefficients; ϵ_j and μ_j are the dielectric permittivities and magnetic permeabilities (which are negative at ω_4); $\Delta k = k_1 + k_3 - k_2 - k_4$; and α_j are the absorption or amplification coefficients. Photon fluxes are given by $S_{2,4}/\hbar\omega_{2,4} = (c/8\pi)|a_{2,4}|^2$. The amplitudes of the fundamental (control) waves E_1 and E_3 are assumed constant along the slab. Transmittance (amplification) at ω_4 is given by the factor $\eta_{4a} = |a_4(0)/a_4(L)|^2$, where L is the slab thickness. Note that the signs in (1) are opposite to those in ordinary media, which is due to the backwardness of the signal wave. The solution to a similar set of equations and its analysis are given in [7,8].

Calculations of the optical constants for embedded NLO centers (driven by the control fields) with account for constructive and destructive quantum interference are performed following the density-matrix technique described in [11]. In our simulations, we used the following representative values for relaxation rates: energy level relaxation rates $\Gamma_n = 20 \times 10^6 \text{ s}^{-1}$, $\Gamma_g = \Gamma_m = 120 \times 10^6 \text{ s}^{-1}$; partial transition probabilities $\gamma_{gt} = 7 \times 10^6 \text{ s}^{-1}$, $\gamma_{gn} = 4 \times 10^6 \text{ s}^{-1}$, $\gamma_{mn} = 5 \times 10^6 \text{ s}^{-1}$, $\gamma_{ml} = 10 \times 10^6 \text{ s}^{-1}$; homogeneous transition half-widths $\Gamma_{lg} = 10^{12} \text{ s}^{-1}$, $\Gamma_{lm} = 1.9 \times 10^{12} \text{ s}^{-1}$, $\Gamma_{ng} = 1.5 \times 10^{12} \text{ s}^{-1}$, $\Gamma_{nm} = 1.8 \times 10^{12} \text{ s}^{-1}$, $\Gamma_{gm} = 5 \times 10^{10} \text{ s}^{-1}$; $\Gamma_{ln} = 10^{10} \text{ s}^{-1}$. We assumed that $\lambda_2 = 756 \text{ nm}$, $\lambda_4 = 480 \text{ nm}$. The results of numerical simulations for the optical coefficients entering equations (1) and (2) are shown in Fig. 2. Here, $\Omega_4 = \omega_4 - \omega_{ml}$; other resonance detunings Ω_j are defined in a similar way. The coupling Rabi frequencies are introduced as $G_1 = E_1 d_{lg}/2\hbar$ and $G_3 = E_3 d_{nm}/2\hbar$. The quantities $\chi_{4,2}$ are effective linear susceptibilities and α_{40} and χ_{40} denote their resonant values when all the driving fields are turned off. Figures 2 (a,b) depict changes in the absorption and amplification coefficients for the signal and idler, respectively, produced by the control fields. (Fig. 2 (b) shows the expanded interval corresponding to nonlinear interference resonances.) Figure 2 (c) displays changes in refractive indices, and Fig. 2 (d) shows wave-vector mismatch. The arrow in Fig. 2 (d) points out the interval of y_4 , roughly between 0.29 and 2.34, with five detunings y_4 for which $\Delta k = 0$ (under the assumption that the host material does not introduce any additional phase mismatch). Figures 2 (e) and 2 (f) show narrow resonances in NLO coupling coefficients, $\gamma_{4,2}^{(3)}$, with the width on the order of other interference resonances. Figure 2 proves the feasibility of manipulating local optical parameters through nonlinear quantum interference induced by the control fields. For the used optical transitions rates, the magnitude of $G \sim 10^{12} \text{ s}^{-1}$ corresponds to control field intensities I of 10 to 100 kW/(0.1mm)².

The optimization of the output signal at $z = 0$ is de-

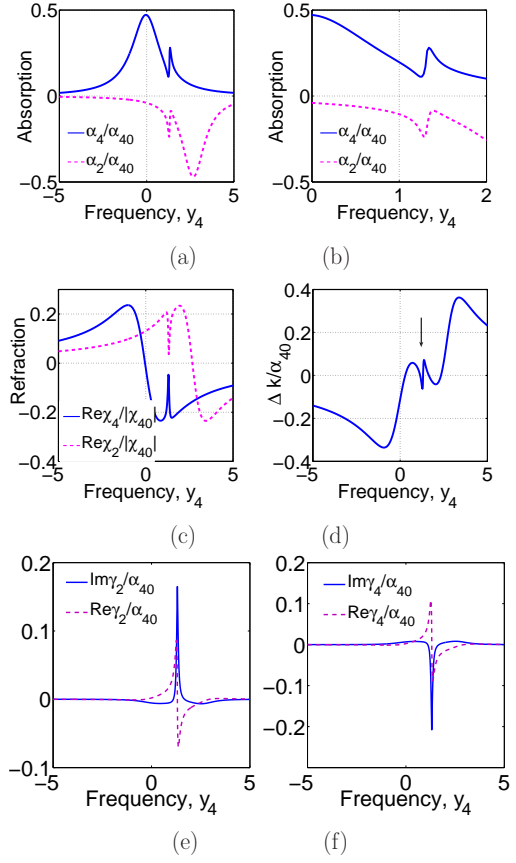


Fig. 2. Nonlinear interference resonances induced by the control fields with $G_1 = G_3 = 50 \text{ GHz}$ and $\Omega_1 = \Omega_3 = 2.5\Gamma_{lg}$. ($y_4 = \Omega_4/\Gamma_{ml}$).

termined by the interplay between absorption, idler gain and FWM, with the later depending on the wave vector mismatch. This is a multi-parameter problem involving sharp resonance dependencies. Results of our numerical analysis of the steady-state solutions [11] to the density matrix equations and Maxwell's equations for the slowly-varying amplitudes in (1) and (2) are shown in Fig. 3. The transmission of the host slab in the NI frequency domain has been set as 10%. Unlike conventional media, the output signal for the waves coupled in the NIM slab through OPA represents a set of distributed feedback-type resonances [7,8]. Such resonant behavior can be observed as a function of the intensity of the fundamental fields, the product of the slab length and the density of NLO centers, and the resonance offsets for the signal and fundamental fields. Figure 3(a) shows these narrow transmission resonances. Here, we introduce the scaled product of the slab length and the density number of embedded centers, L/L_{ra} , through the resonance absorption length, $L_{ra} = \alpha_{40}^{-1}$. Figure 3(b) displays the second peak in Fig. 3(a) with greater detail. It shows that the transparency window is on the scale of the narrowest (Raman, in this case) transition half-width, as a function of detuning, and the resonant

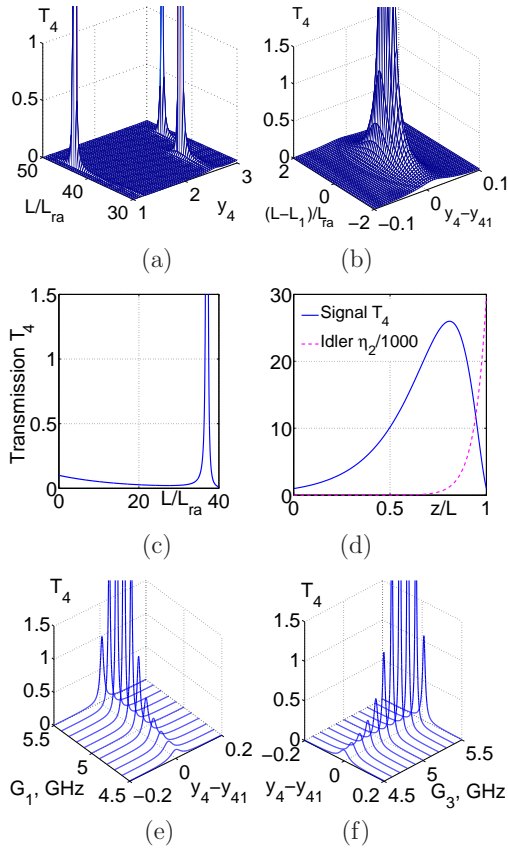


Fig. 3. Laser-induced transmission resonances in the NI frequency domain. $\Omega_1 = \Omega_3 = 2.5 \cdot \Gamma_{lg}$. (a)-(d): $G_1 = G_3 = 50$ GHz. (a)-(c), (e)-(f): $z = 0$. (c)-(d): $y_4 = y_{41} \equiv 2.5266$. (c): maximum is at $\alpha_{40}L = L/L_{ra} = L_1/L_{ra} \equiv 37.02$, $T_4 \approx 1$ at $L/L_{ra} = 36.52$. (d): intensity distribution inside the slab: signal – solid line, idler – dashed line; $L/L_{ra} = 36.52$. (e) and (f): $L = L_1$. (e): $G_3 = 50$ GHz. (f): $G_1 = 50$ GHz.

absorption length, as a function of length. Figure 3(c) depicts the same dependence at the optimum resonance offset. Figure 3(d) shows that the intensity of the signal inside the slab may significantly exceed its output value at $z=0$, which depends on the ratio of the OPA and absorption rates. Here, $\eta_2(z) = |a_2(z)/a_{4L}|^2$. Remarkably, the transparency and amplification occur in the frequency range where magnitudes of $|\gamma_{4,2}|$ are substantially less than their resonant values. Figures 3(e) and 3(f) display similar resonance dependence on the strength of the control fields. The amplification in the maximums in Fig. 3 reaches many orders of magnitude, which indicates the *feasibility of oscillations* and, hence, the generation of counter-propagating left-handed signal and right-handed idler photons. It is known that even small amplification per unit length may lead to lasing provided that there is a high-quality cavity (or feedback resonance) which effectively increases the distance over which the amplification occurs.

According to Fig. 3(c), characteristic values of $\alpha_{40}L \sim 10$ are required to ensure the transparency and gain. Assuming $\sigma_{40} \sim 10^{-16}$ cm² for the resonance absorption cross-section, which is typical for dye molecules, and $N \sim 10^{19}$ cm⁻³ for the density of molecules, we obtain that $\alpha_{40} \sim 10^3$ to 10^4 cm⁻¹, and the required slab thickness in the range of $L \sim 10$ to 100 μ m. At these values, the contribution of the non-linear centers in the refractive index is estimated as $\Delta n < 0.5(\lambda/4\pi)\alpha_{40} \sim 10^{-2} - 10^{-3}$, which essentially does not change the linear negative refractive index.

In conclusion, we propose the compensation of losses in strongly absorbing NIMs through embedded tailored optical nonlinearities. Such a possibility is shown with a realistic numerical model. We have studied the resonant FWM-based OPA in such composite metamaterials with a negative refractive index at the frequency of the signal and a positive index for all other coupled waves. The strong nonlinear optical response of the composite is primarily determined by the embedded four-level nonlinear centers and, hence, can be adjusted independently. In addition, we have shown the possibility of quantum control of the local optical parameters, which employs constructive and destructive quantum interference tailored by two auxiliary control fields. Frequency-tunable transparency windows in the negative-index frequency domain, cavity-free generation of entangled counter-propagating photons, and the feasibility of quantum switching in NIMs have been shown.

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